Interpretation of SEAC⁴RS Aerosol Observations over the Southeast US with the GEOS-Chem Chemical Transport Model

Patrick Kim[1], Daniel Jacob[1], Jenny Fisher[2], Jose Jimenez[3], John Hair[4], Katherine Travis[5], Lei Zhu[5], Karen Yu[5], and the SEAC⁴RS Science Team

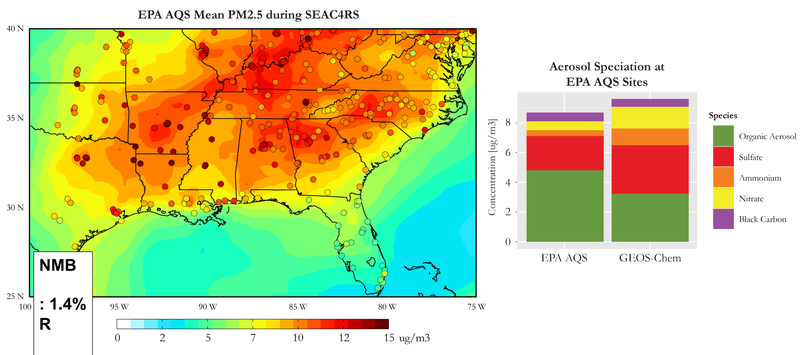
[1]Harvard University, Department of Earth and Planetary Science | [2]University of Wollongong | [3]University of Colorado-Boulder

[4]NASA Langley Research Center | [5]Harvard University, School of Engineering & Applied Science

Motivation

Satellite aerosol optical depth (AOD) has been used extensively to quantify aerosol sources and as a proxy for surface air quality [Dubovik et al. 2008; van Donkelaar et al. 2013]. However, this requires independent information on aerosol composition, chemistry, and vertical distribution, typically provided by a chemical transport model (CTM). The Southeast US presents a complex environment where we can test and improve our understanding of these external factors, due to high natural emissions in the region coupled with a rapid decline in anthropogenic pollution. In addition, satellite observations show a pronounced regional summer AOD maximum [Goldstein et al., 2009], which has been attributed to biogenic secondary organic aerosol with an unidentified free tropospheric source [Ford and Heald, 2013]. Understanding this seasonal aerosol feature will be important for surface aerosol characterization using satellite retrievals. Here we present a preliminary evaluation of a high resolution version of the GEOS-Chem CTM with the SEAC⁴RS aircraft and ground station measurements to test our understanding of aerosol sources and fate in the Southeast US, working towards the goal of improving the interpretation of satellite AOD data.

Ground Station



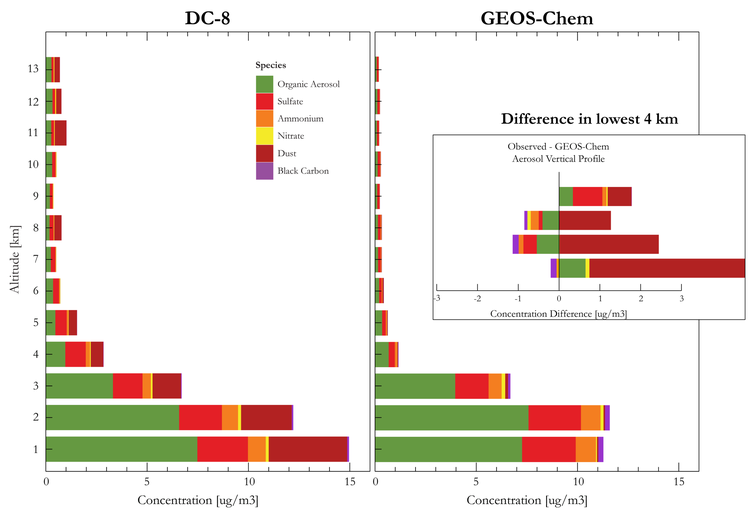
Mean surface $PM_{2.5}$ concentration at EPA Air Quality System (AQS) stations during SEAC⁴RS. GEOS-Chem $PM_{2.5}$ is shown as background. (Right) Measured and simulated aerosol speciation across all EPA AQS stations in the Southeast US domain shown above.

- Observed mean $PM_{2.5}$ concentration is 11.2 ug/m^3 with max over LA/MS/AR. GEOS-Chem captures magnitude but not variability of the surface station data.
- AOD decreases across all AERONET sites in late Sept., but large variability among stations.

(Left) Timeseries of median level 1.5 AERONET AOD across all stations and (Bottom) at representative sites in the Southeast US. Standard deviation is shown to highlight daily variability among sites. Also shown is the GEOS-Chem simulated AOD and contribution from sulfate and organic aerosols.

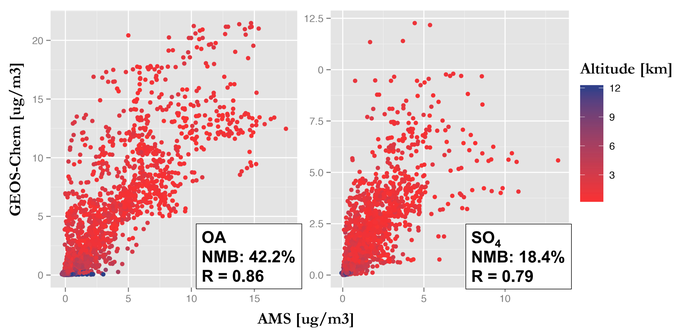
We thank B. Holben & the AERONET team for establishing the regional sun photometer network during SEAC⁴RS.

In-Situ

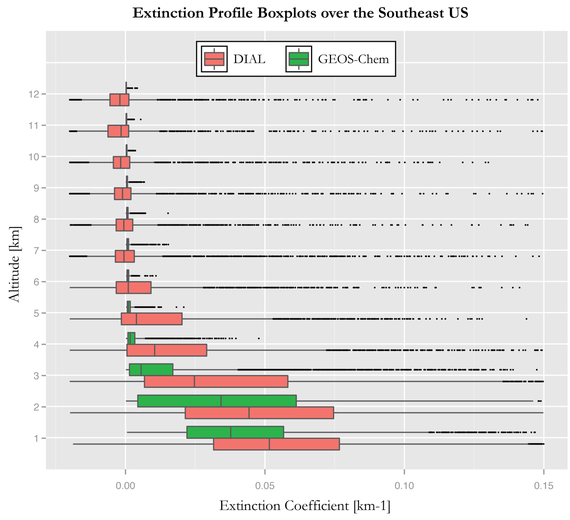


Median aerosol vertical profiles measured aboard the DC-8 and simulated by GEOS-Chem at the plane flight time. Organic aerosol, sulfate, nitrate, and ammonium from the AMS (J. Jimenez, CU Boulder), dust derived from Ca^{2+} and Na^{+} from SAGA (J. Dibb, UNH), and black carbon from HDSP2 (R. Gao, NOAA). Data and model are binned to 1 km resolution.

- Organic aerosol is the single largest contributor to aerosol concentration in the boundary layer
- Observed and simulated total aerosol concentrations in the 0-1 km layer are consistent with EPA AQS station data, when dust concentrations are excluded. However, the dust concentrations dominate the observed-simulated differences shown inset above and could explain the extinction underestimate shown in the Remote Sensing panel. Further investigation is required.
- Strong correlation of simulated concentrations with individual aerosol species (scatterplot of AMS observed and simulated OA and sulfate below – SOA parameterization following Hodzic & Jimenez [2011]).

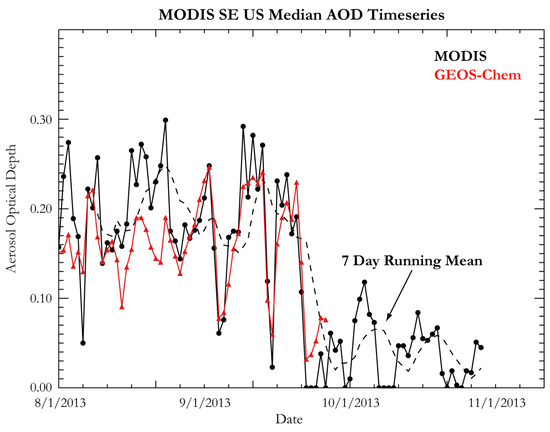


Remote Sensing



Median aerosol extinction profiles (550 nm) measured aboard the DC-8 (DIAL, J. Hair, NASA Langley) and simulated by GEOS-Chem. Data and model are binned to 1 km resolution.

- Extinction is generally low above 5 km, though plumes are common.
- GEOS-Chem underestimates DIAL extinction between 0 – 4 km. The integrated difference in extinction between 0 – 4 km is consistent with the underestimate in AOD shown in the AERONET data (Ground Station panel), 4STAR (not shown), and MODIS (below).
- Seasonal transition to lower AOD in the SE US seen at AERONET stations and in MODIS data. Simulated AOD decline associated with decrease in biogenic emissions and PAR.



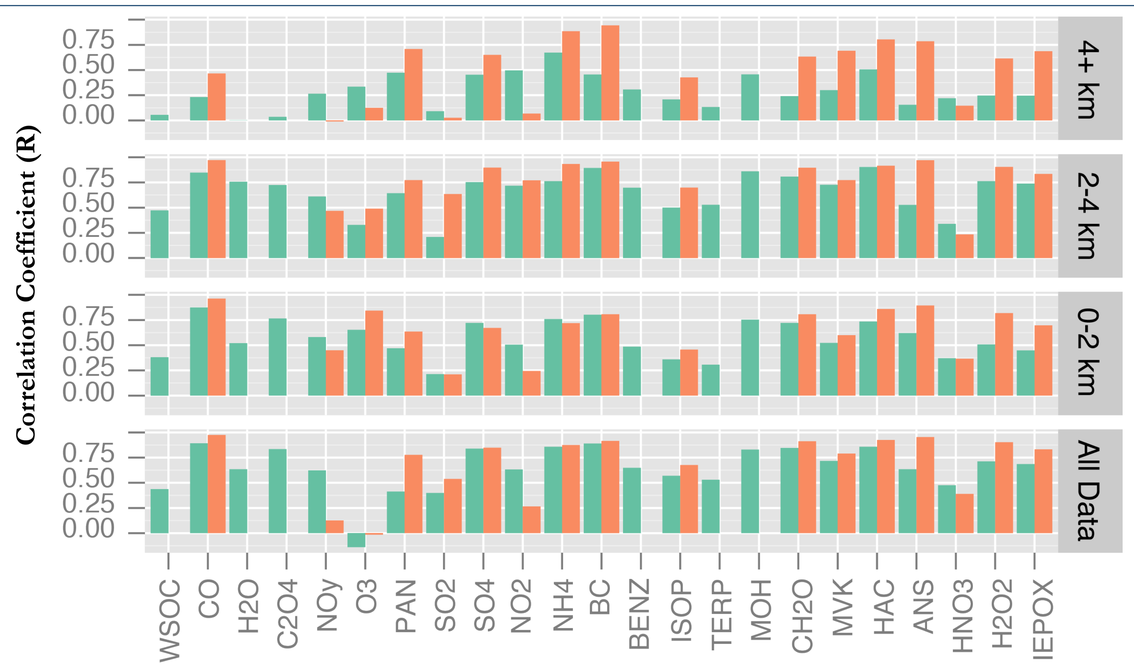
Daily Median SE US AOD (region defined here as 30-40N, 80-100W) retrieved by MODIS and sampled at the satellite overpass time by GEOS-Chem.

The Level 3 MODIS data is filtered for sensor saturation and clouds as in Ford and Heald [2013].

There is a sharp decrease in the regional median AOD in late September.

OA Correlation Analysis

- We explore the key processes that lead to organic aerosol formation by examining its correlations with other chemical variables.
- OA correlations with CO are high in the boundary layer but low in the free troposphere, suggesting aerosol scavenging during uplift.
- We use WSOC as a tracer for secondary organic aerosol (SOA). The observed OA-SOA correlation is moderate in the boundary layer and drops off sharply in the free troposphere.
- A strong candidate for the formation of SOA is through aqueous processing. This is supported by strong correlations of OA with water vapor and oxalate, as well as a significant correlation to sulfate.
- The parameterization for SOA production used in GEOS-Chem does not include aqueous processing. The model ability to capture OA variability decreases as a function of height (see In-Situ Aerosol panel).
- Strong correlation of OA to black carbon at all altitudes. Coupled with benzene, and because we filter for fire plumes, this suggests there is strong anthropogenic influence on OA formation in the Southeast US.
- Clear influence of biogenic precursors and oxidation chemistry through correlations with isoprene and monoterpenes, as well as both high and low NO_x oxidation products.
- In future work, we will perform more rigorous statistical model building and factor analysis.



LEGEND: DC8 GEOS-Chem

Correlation of AMS OA and PILS water soluble organic carbon (WSOC) with other measured species aboard the DC8. The GEOS-Chem correlations are shown for simulated OA to other variables in the model, sampled at the aircraft position and time. Total simulated OA is shown in the comparison with the AMS, while only SOA is shown in the comparison with WSOC. The OA relationships are shown for different altitude ranges and have been filtered for fire plumes.

We thank R. Weber (PILS WSOC, Georgia Tech), G. Diskin (DACOM CO and DLH H_2O, NASA LaRC), J. Dibb (SAGA C_2O_4, UNH), T. Ryerson (ESRL NO_y and O_3), G. Huey (CIMS PAN and SO_2, Georgia Tech), R. Gao (HDSP2 BC, NOAA), Don Blake (WAS Benzene, UC Irvine), A. Wisthaler (PTR-MS Isoprene, Monoterpenes, MVK + MACR, and Methanol, University of Innsbruck), T. Hanisco (LIF CH_2O, NASA GSFC), Ron Cohen (TDLIF ANs, UC Berkeley) and Paul Wennberg (CIMS HAC, HNO_3, H_2O_2, and IEPOX, Caltech) for making the preliminary data available.

References:
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kim68@fas.harvard.edu